A Perspective on Autonomous Experimentation and Discovery

Marcus Michael Noack







What is Autonomous Data <u>Acquisition</u>

> <u>CAMERA Workshop on AE</u> and Main Takeaways

> > <u>Basic</u> <u>Gaussian-Process-Driven</u> <u>Autonomous Data</u> <u>Acquisition</u>



<u>Mathematical</u> <u>Optimization for</u> <u>AE and ML</u>



Bringing Autonomous Discovery to the <u>Community</u>



What is Autonomous Data Acquisition



	Random	Scanning	Intuition
Inefficient	Prev. data not	Prev. data not	Not autonomous/
	used/Non-optimal	used/Non-optimal	human attention
	measurements	measurements	required
Uninformative	No metric for	No metric for	No metric for
	quality	quality	quality
Biased			Relies on past experience Lack of reproducibility







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<u>CAMERA Workshop on</u> <u>AE and Main Takeaways</u>



Autonomous Discovery for Science and Engineering

A three-day workshop for sharing recent developments in autonomous methods, sponsored by CAMERA — The Center for Advanced Mathematics for Energy Research Applications

Dates: April 20th - 22nd, 2021

Methods and Algorithms: Gaussian Processes, Neural Networks, Reinforcement Learning, New Math, Optimization, Data Analytics and Infrastructure, UQ

Tutorials: gpCAM, Summit, CamLink, Escalate, Bluesky, Atinary SDLabs, DataFed, Cameo, AtomAI, ART

Applications: Microscopy, Spectroscopy, X-ray Scattering, Neutron Scattering, Autonomous Synthesis & Materials Discovery, Robotics & Remote Access



DOI: https://doi.org/10.2172/1818491

https://autonomous-discovery.lbl.gov/

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The Organizing Committee





Simon Billinge, Columbia U.



Petrus Zwart, LBNL



Apurva Mehta, SLAC



Nicholas Schwarz, ANL



Martin Boehm, ILL



Daniela Ushizima, LBNL







James Sethian, UCB

Kevin Yager, BNL



Alex Hexemer, LBNL



Aaron Gilad Kusne, NIST



Bobby Sumpter, ORNL

B. Reeja Jayan, CMU



Sergei Kalinin, ORNL





What is your background? 512 responses



Computer Science Chemical Engineering Chemical Engineering Mechanical Engineering Engineering Computer science engineering Electrical Engineering

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- Computational Biology
- Radiation Therapy
- Lead a Data Science team at PNNL b...
- Computer Science, Data Science
- X-ray science
- Bioengineering

▲ 3/6 ▼

- computer science
- Chemiclal Engineer

▲ 4/6 ▼

▲ 5/6 ▼

- computational mechanics
- Application to IT Infrastructure Engineering in support of Joint Genome Institute..
- Computer Engineering
- Optimization, Chemical engineering, machine lerning
- social sciences, ethics

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Takeaways and Achievements: Methods/Algorithms



Gaussian and

other stochastic processes

Modelling



Reinforcement Learning

Take-Home Message 1: Gaussian processes and reinforcement learning are the most popular techniques for control, in different data regimes. Analysis



Take-Home Message 2: The data-analysis step is more and more often done automatically data-science tools (PCA, NN, Clustering,...).

Hardware/Robotics



Take-Home Message 3: Instrument systems are increasingly built with automation in mind.

Training and Decision-Making



Take-Home Message 4: Efficient mathematical optimization under uncertainty and subject to constraints has come far but remains a challenge.

Communication Infrastructure



Take-Home Message 5: Data-management systems are emerging using lower-level tool (control, optimization, ...) to allow for standardization.

Takeaways and Achievements: Application



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Challenges

The Role of Co-Design: Much of the work is performed through co-design teams, bringing together needed expertise. The work has aspects of theory, modeling, algorithm design, data analysis, workflow, and software engineering.

Integrating Across Required Expertise: Teams (or in some cases, individuals) working in autonomous design often take on all the required roles, which requires a large breadth of expertise: it is challenging for a team to excel in all the necessary aspects.

Sharing Developments: There are significant opportunities to share advances across autonomous efforts. However, there is often inconsistent nomenclature and problem formulation.

Workflow and Infrastructure: For the most part, individual efforts center around homegrown workflows and infrastructures. There are opportunities to build work tools and infrastructures that can be shared.

Software: Understandably, many of the efforts described within are aimed at solving a particular set of scientific problems, and the emphasis is not on generalizable software. Openly-available software that is well-documented and properly maintained would be a step forward.

Shared Testbeds and Reproducible Research: It is challenging to cross-test different algorithms and methodologies with common accessible (FAIR) datasets with maintained standards.

Data: Data != data. The structure of data has to be discussed before data taking and ML applications.

Compute Resources: The field requires a new kind of compute-resource allocation, which keeps resources available throughout the experiment.





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Gaussian Process Regression in a Nutshell

$$p(\mathbf{f}) = \frac{1}{2\pi^{d/2}} |\Sigma|^{-1/2} \exp\left[-\frac{1}{2}(\mathbf{f} - \boldsymbol{\mu})^T \Sigma^{-1}(\mathbf{f} - \boldsymbol{\mu})\right]$$

$$\mathcal{H} = \{f(\mathbf{x}) : f(\mathbf{x}) = \sum_{i}^{N} \alpha_i k(\mathbf{x}_i, \mathbf{x}), \forall \mathbf{a} \in \mathcal{R}^N, \mathbf{x} \in \mathcal{R}^n\}$$

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Our Very First Experiment: A Nanoparticle Stain Mapping Experiment Facility: NSLS2, CFN @ BNL | Technique: SAXS | Achievement: Commissioning experiment





Center for Functional Nanomaterials Brookhaven National Laboratory

Autonomous SAXS Exploration of Nanoscale Ordering in a Blade-Coated Polymer-Grafted Nanorod Film

Facility: AFRL and NSLS II | Technique: SAXS | Achievement: 15% of data required, higher resolution in areas of interest



Autonomous Steering of ARPES Data Acquisition Facilities: ALS @ LBNL | Technique: ARPES | Achievement: 12% of data required







K-Means-Driven Gaussian Process Data Collection for Angle-Resolved Photoemission Spectroscopy Charles N. Melton, Marcus M. Noack, Taisuke Ohta, Thomas E. Beechem, Jeremy Robinson, Xiaotian Zhang, Aaron Bostwick, Chris Jozwiak, Roland J. Koch,

Petrus H. Zwart, Alexander Hexemer, and Eli Rotenberg

Autonomous Control of Synchrotron Infrared Spectroscopy Facility: ALS @ LBNL | Technique: IR Spec. Micr. | Achievement: ~5% of data required, collected in ~10% of the time, materials targeted





Hoi-Ying Holman, Petrus Zwart, Liang Chen, Steven Lee



1 hour

9 hours

9.8K sample points











Autonomous Scanning Tunneling Spectroscopy

Facility: Molecular Foundry @ LBNL | Technique: STS Microscopy | Achievement: ~4% of data required, ~35 hrs vs ~1 mo acq. time





Thomas et al., arxiv:2110.03351 (2021)

The Power of the RKHS: Domain–Informed Symmetry Constraints — Six–Fold Symmetry

$$k(\mathbf{x}_i, \mathbf{x}_j) = \frac{1}{36} \sum_{\phi} \sum_{\theta} \tilde{k}(\mathcal{R}_{\phi} \mathbf{x}_i, \mathcal{R}_{\theta} \mathbf{x}_j)$$







Martin Boehm Paolo Mutti Tobias Weber

Physics Knowledge in the Form of Periodicity for X–Ray Scattering Facility: NIST and NSLS II | Technique: SAXS/GISAXS | Achievement: Use of non-stationary kernels to learn and exploit local characteristics





Physics – Aware Prediction of Lattice Thermal Conductivity of Alloys Facility: SLAC @ Stanford | Technique: Diffusivity, Heat Capacity and Density Measurements | Achievement: Physics-informed GP-driven steering

Data-Driven GPR



Physics-Based Model



Suchismita Sarker, Apurva Mehta

Physics-Aware GPR



Targeted Autonomous Neutron Scattering Facility: ILL, France | Technique: Inelastic Neutron Scattering | Achievement: More efficient exploration, experiment time decreased from several days to one night



Martin Boehm Paolo Mutti Tobias Weber





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The Traditional Training/Optimization Workflow needs a Large Number of Function Evaluations and Blocks the Main Thread

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Minimizing Number of Function Evaluations: Asynchronous Distributed Training



Optimization of the Log-Likelihood and Acquisition Functions with HGDL

Using DASK, pytorch and GPUs for High Performance Asynchronous Distributed Training



$$\log(L(\mathcal{D};\phi,\mu)) = -\frac{1}{2}(\mathbf{y}-\boldsymbol{\mu}(\mathbf{x}))^T(\mathbf{K}(\phi)+\mathbf{I}_e)^{-1}(\mathbf{y}-\boldsymbol{\mu}(\mathbf{x})) - \frac{1}{2}\log(|\mathbf{K}(\phi)+\mathbf{I}_e|)$$



HGDL leads to:

- 1. a set of different interpretations of the data
- 2. a set of optimal measurements
- 3. HPC readiness of training and prediction
- 4. Asynchronous training



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<u>Bringing</u> <u>Autonomous</u> <u>Discovery to the</u> <u>Community</u>

HGDL: As<mark>ynch.</mark> Distributed Optimizer

fvGP: A flexible multi-task Gaussian process tool



pip install gpcam

```
def instrument(data):
    for entry in data:
        entry["value"] = np.sin(np.linalg.norm(entry["position"]))
    return data
```

More information: gpcam.lbl.gov



Richard Vaia

Kit Windows-Yule •

- Marina Ganeva
- Mario Parente





A Perspective on Autonomous Experimentation and Discovery

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Questions?

